

Acoustic Attenuation in Glasses and its Relation with the Boson Peak

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A theory for the vibrational dynamics in disordered solids [W. Schirmacher, *Europhys. Lett.* **73**, 892 (2006)], based on the random spatial variation of the shear modulus, has been applied to determine the wave vector (k) dependence of the Brillouin peak position (Ω_k) and width (Γ_k), as well as the density of vibrational states [$g(\omega)$], in disordered systems. As a result, we give a firm theoretical ground to the ubiquitous k^2 dependence of Γ_k observed in glasses. Moreover, we derive a quantitative relation between the excess of the density of states (the boson peak) and Γ_k , two quantities that were not considered related before. The successful comparison of this relation with the outcome of experiments and numerical simulations gives further support to the theory.

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The most striking differences between glasses and crystals, at a macroscopic level, concern the thermal properties. At few tens of Kelvin, the specific heat of glasses exhibits an excess over the Debye expectation, which, in a $C(T)/T^3$ vs. T plot, appears as a characteristic maximum. Similarly, at low- T the thermal conductivity increases as T^2 with increasing T and, in the same temperature range where $C(T)/T^3$ has a maximum, exhibits a plateau. While the first observation can be ascribed to the presence of an excess of states over the Debye density of states (DOS) $g_D(\omega)$ (the "boson peak", i.e., the peak observed in $g(\omega)/\omega^2$ vs. ω), the second one was until recently not understood at all. Both anomalies appear to be strongly affected by the characteristics of the normal modes of vibrations in the THz frequency region. The nature of these modes has been the subject-matter of a very intense and controversial debate in the literature since many decades. The issue has been recently revitalized thanks to new neutron, X-ray, and other inelastic scattering experiments [1–10], computer simulations [11–17], and analytical theory [18–27].

The boson peak shows up in a frequency range where the broadening of the acoustic excitations becomes of the order of magnitude of the resonance frequency, thus indicating a possible mode localization. This observation lead different authors to hypothesize a relation between the position of the boson peak and the existence of localized vibrations. Acoustic waves that become Anderson localized, indeed, could produce the plateau in the thermal conductivity. Following this idea, investigations of (Anderson-)localization properties of waves in disordered systems based on simulations [12], model calculations [19,20], and field-theoretical techniques [28] have shown that Anderson-localized states in disordered media do actually occur, but in a much higher frequency range (near the upper band edge) than the boson peak frequency.

So the question is still open: what is the very nature of the states near and above the boson peak frequency? As these states are neither really propagating nor localized,

Feldman and co-workers [12] suggested calling them "diffusons" (they behave like diffusing light in milky glass). In this regime, however, the resonance frequency Ω_k still exhibits a linear dispersion with the wave vector k , albeit with an apparent sound velocity being somewhat larger than the ultrasonic speed [16]. More interesting, in this frequency range, the width Γ_k of the excitations, a quantity proportional to the sound attenuation coefficient, shows a k^2 dependence in most of the investigated materials [6], whereas at lower frequencies the width appears to have a stronger k dependence [9]. These findings are still awaiting a theoretical explanation.

In this Letter we exploit a recently developed theory of vibrational excitations in disordered elastic media [29] and apply it to the calculation of the dynamic structure factor. This theory is based on the model assumption that the disorder leads to microscopic random spatial fluctuations of the transverse elastic constant (shear modulus) [30]. As in similar, more schematic approaches [20,23,25] the excess DOS has been shown to arise from a band of disorder-induced irregular vibrational states, the onset of which approaches lower frequencies as the disorder is increased. Within this framework it was possible to formulate a theory for the energy diffusivity, which gave the first explanation of the plateau of the temperature dependent thermal conductivity and its relation to the excess DOS. When applied to the calculation of the shape of the Brillouin resonance, this theory *explains* (i) the ubiquitous k^2 dependence of Γ_k and (ii) the observed increase of the apparent sound velocity with frequency [31]. Moreover, the theory *predicts* (iii) that the excess of vibrational states, if properly normalized with the Debye DOS, takes almost a universal value and (iv) the existence of a quantitative relationship between the excess over the Debye DOS and the width of the Brillouin line. The latter, previously unexpected, relation is shown to agree with the findings of experimental inelastic scattering and computer simulation experiments.

We now shortly summarize the theory. We consider an elastic medium with a mass density m_0 , shear modulus G , bulk modulus $K = \lambda + \frac{2}{3}G$ (λ = longitudinal Lamé constant). These elastic constants are related to the longitudinal and transverse local sound velocities as $c_T^2 = G/m_0$; $c_L^2 = (K + \frac{4}{3}G)/m_0 = (\lambda + 2G)/m_0$. We now assume that the shear modulus (and not λ , which is set to a constant value λ_0) exhibits a random spatial variation: $G(\mathbf{r}) = G_0[1 + \Delta\tilde{G}(\mathbf{r})]$. The random function $\Delta\tilde{G}(\mathbf{r})$ is supposed to be Gaussian distributed around its average G_0 with a variance $\propto \gamma_G$. The parameter γ_G describes the “degree of disorder” of the system. We emphasize that this phenomenological model may be adequate both for a topologically disordered system (glass) or a disordered crystal. Using standard field-theoretic techniques [25,28,29,32] the self-consistent Born approximation (SCBA) for the (complex) self-energy function $\Sigma(\omega) = \Sigma'(\omega) + i\Sigma''(\omega)$ has been derived [29], resulting in the set of equations

$$\begin{aligned}\Sigma(\omega) &= \gamma_G \Sigma_{k < k_D} [\chi_L(k, \omega) + \chi_T(k, \omega)], \\ \chi_L(k, \omega) &= k^2 \{-\omega^2 + k^2 [c_{L,0}^2 - 2\Sigma(\omega)]\}^{-1}, \\ \chi_T(k, \omega) &= k^2 \{-\omega^2 + k^2 [c_{T,0}^2 - \Sigma(\omega)]\}^{-1}.\end{aligned}\quad (1)$$

Here χ_L (χ_T) are the longitudinal and transverse dynamic susceptibilities, respectively, and $c_{L,0}$, $c_{T,0}$ are the sound velocities of a system without disorder. The sum over k indicates integration up to the Debye cutoff, $k_D = [6\pi^2 N/V]^{1/3}$, according to the identification $\Sigma_{k < k_D} \rightarrow (3/k_D^3) \int_0^{k_D} k^2 dk$. The SCBA is the simplest form of an effective-medium theory for the disorder [19,20,23], in which the disorder effects lead to a frequency dependent modification of the sound velocities (“complex acoustic indices of refraction”) by the function $\Sigma(\omega)$. The real part $\Sigma'(\omega)$ causes a renormalization and dispersion of the sound velocities according to $c_L(\omega)^2 = c_{L,0}^2 - 2\Sigma'(\omega)$ and $c_T(\omega)^2 = c_{T,0}^2 - \Sigma'(\omega)$, whereas its imaginary part describes the sound attenuation (see below). Apart from the natural length and frequency scales k_D^{-1} and $\omega_D = c_D k_D$ [33] there are only two nontrivial parameters in this theory, namely, the degree of disorder γ_G and the ratio of the renormalized sound velocities c_L/c_T [33].

From Eq. (1), the DOS can be calculated as

$$g(\omega) = (2\omega/3\pi) \Sigma_{k < k_D} \frac{1}{k^2} \text{Im}\{\chi_L(k, \omega) + 2\chi_T(k, \omega)\}.\quad (2)$$

As in other theories of a harmonic solid with quenched disorder [20,23,26], the system becomes unstable if the disorder exceeds a critical value γ_G^c , which—in the present case—slightly depends on the ratio of sound velocities and ranges, for example, from $\gamma_G^c = 0.1666$ for $c_L^2/c_T^2 = 2$ to $\gamma_G^c = 0.227$ for $c_L^2/c_T^2 = 6$. It has been demonstrated [20,23,25,26,29], that the quenched disorder produces an excess over the Debye DOS, the offset of which approaches lower and lower frequencies as $\gamma_G \rightarrow \gamma_G^c$.

In Fig. 1 we report the *reduced* DOS $g(\omega)/g_D(\omega)$ (i.e., the usual boson peak representation) and the *reduced excess* DOS $[g(\omega)/g_D(\omega)] - 1$ against ω/ω_D for several values of the separation parameter $\epsilon = 1 - \gamma_G/\gamma_G^c$. It is remarkable that the value of the normalized excess does not exceed the value 1, which it takes for small enough values of ϵ . It is also remarkable that the excess essentially does not depend on the ratio of the sound velocities. Moreover, the excess is significantly different from zero *only above* a certain frequency threshold (onset frequency), below which the DOS basically coincides with the Debye expectation. The excess DOS turns out to vanish as ω^4 for $\omega \rightarrow 0$.

We now turn the attention to the dynamical structure factor, $S(k, \omega)$, which can be measured by inelastic neutron, x-ray, and light scattering:

$$\begin{aligned}S(k, \omega) &= \frac{1}{\pi} [n(\omega) + 1] \text{Im}\{\chi_L(k, \omega)\} \\ &= \frac{1}{\pi} [n(\omega) + 1] 2k^4 \Sigma''(\omega) \{ [k^2 c_L(\omega)^2 - \omega^2]^2 \\ &\quad + 4k^4 \Sigma''(\omega)^2 \}^{-1} \\ &\approx \frac{1}{\pi} [n(\omega) + 1] \frac{k^2}{2\omega} \\ &\quad \times \frac{k^2 \Sigma''(\omega)/\omega}{[c_L(\omega)k - \omega]^2 + [k^2 \Sigma''(\omega)/\omega]^2}.\end{aligned}\quad (3)$$

From this equation we can read off the width of the Brillouin line (full width at half maximum) as

$$\Gamma(\omega) \approx 2k^2 \Sigma''(\omega)/\omega \approx \frac{2}{c_L(\omega)^2} \omega \Sigma''(\omega).\quad (4)$$

As the “disorder function” $\Sigma''(\omega)$ enters also into the DOS we find from (2) for $\Sigma''(\omega) < c_{L,T}(\omega)^2$ the following ap-

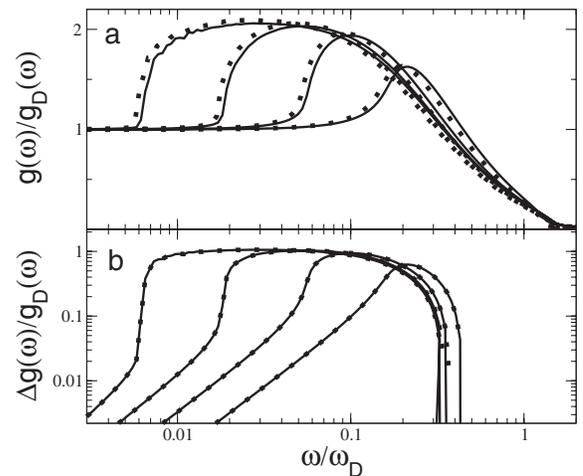


FIG. 1. (a) Reduced DOS $[g(\omega)/g_D(\omega)]$ for ($\epsilon = 1 - \gamma_G/\gamma_G^c = 0.0001, 0.001, 0.01, \text{ and } 0.1$) (from left to right) and for $(c_L/c_T)^2 = 2$ (full lines) and $(c_L/c_T)^2 = 6$ (dots). (b) Reduced excess DOS $\Delta g(\omega)/g_D(\omega)$ for the same parameters [33].

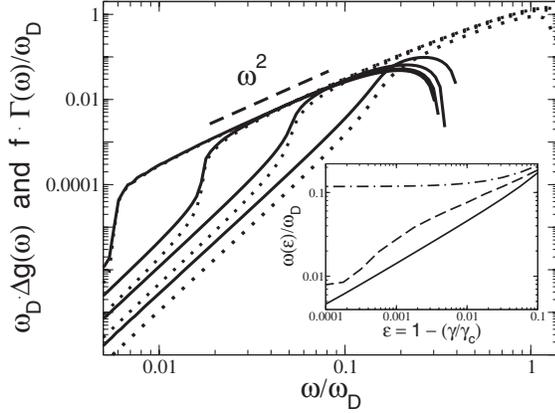


FIG. 2. Excess DOS $g(\omega) - g_D(\omega)$ for the same ϵ values as in Fig. 1 and for $(c_L/c_T)^2 = 6$ (full lines) together with the Brillouin linewidth $f(c_L, c_T)\Gamma(\omega)/\omega_D = 2\omega f(c_L, c_T)\Sigma''(\omega)/\omega_D c_L^2$ (dotted lines). Inset: Onset frequency (full line), boson peak frequency (dashed line) and Ioffe-Regel frequency $\omega_{IR} = c_L(\omega)k/\pi$ (dash-dotted line) as a function of the parameter $\epsilon = 1 - \gamma_G/\gamma_c$.

proximate relation

$$\omega_D \Delta g(\omega) = \omega_D [g(\omega) - g_D(\omega)] = f(c_L, c_T) \frac{\Gamma(\omega)}{\omega_D}, \quad (5)$$

with

$$f(c_L, c_T) = \frac{2}{\pi} (c_D/c_L)^2 [1 + (c_L/c_T)^4]. \quad (6)$$

To check the validity of the approximation made in deriving Eq. (5), in Fig. 2 we compare the excess DOS with the Brillouin linewidth, multiplied with the factor $f(c_L, c_T)$. We see that within our theory the approximate relationship given in Eq. (5) holds, particularly in the regime where both quantities follow approximately an ω^2 dependence.

Equation (5) represents the main result of the present work. It tells us that the Brillouin width $\Gamma(\omega)$ is proportional to ω^2 [and thus to k^2 recalling the slightly changing value of $c_L(\omega)$] in the whole frequency region covered by the boson peak, and that it turns to a ω^4 behavior only

below the onset frequency. The onset frequency, that must not be confused with the boson peak frequency ω_{BP} (see inset of Fig. 2), lies well below ω_{BP} , it is located at the low frequency edge of the boson peak (i.e., at that frequency where the excess DOS vanishes) and it tends to zero as $\epsilon \rightarrow 0$. Equation (5) also gives a quantitative prediction: it establishes a relation between the absolute value of the excess DOS and the Brillouin peak width. The material dependent physical quantities entering into this relation are only macroscopic properties, as sound velocities and density, that can be determined independently.

In order to test the theoretical prediction manifested in Eq. (5) we have collected a number of experimental and numerical simulation data of the vibrational DOS and the Brillouin line width, as well as sound velocity data (Table I). A complete set of data, to our knowledge, only exists in the case of the numerical simulation of Lennard-Jones particles and for a model of silica glass, and for the experimental cases of silica, germania, selenium, glycerol and *o*-terphenyl.

The first test is that of Fig. 1(a) from which we expect that the quantity $\rho = [g(\omega)/g_D(\omega)]_{\max} = A/A_D$ (where A, A_D are the coefficients of the measured and Debye ω^2 law in the boson peak regime) is equal to 2. A systematic variation upwards is observed, but the overall set of data is not far from this value [45]. Exceptions are the case of silica, where the presence of specific localized modes (tetrahedra rotation) are well documented, and that of selenium, where the ratio is low, most likely because the boson peak is not well developed, indicating a large value of ϵ .

The second test involves the quantity $R_{\text{exp}} = \Delta g(\omega)/\Gamma(\omega) = (A - A_D)/B$, which is the ratio of the excess DOS to the Brillouin width as determined by the experiments, while R_{Th} is the same ratio as determined from the theoretical prediction, i.e., $R_{\text{Th}} = f(c_L, c_T)/\omega_D^2$. The values of the two quantities are very similar in all the cases investigated, the maximum deviation, observed once more for the cases of silica and selenium, being within a factor two. The column $Err = |R_{\text{Exp}} - R_{\text{Th}}|/[(R_{\text{Exp}} + R_{\text{Th}})/2]$ reports the percentage of deviation between the

TABLE I. Data taken from the literature together with quantities derived from them: atomic volume V/N , longitudinal and transverse sound velocities $c_{L,T}$; coefficients A, A_D of the ω^2 dependence of the measured and Debye DOS; coefficient B entering into the frequency dependence of the Brillouin width, $\Gamma(\omega) = B\omega^2$; Debye wave number $k_D = [6\pi^2 N/V]^{1/3}$, Debye sound velocity c_D [33], and frequency $\omega_D = c_D k_D$; $\rho (= A/A_D)$, $R_{\text{exp}} = (A - A_D)/B$, $R_{\text{Th}} = f(c_L, c_T)/\omega_D^2$, $Err = |R_{\text{Exp}} - R_{\text{Th}}|/[(R_{\text{Exp}} + R_{\text{Th}})/2]$.

Material	V/N \AA^3	c_L m/s	c_T m/s	A ps ³	B ps	k_D \AA^{-1}	c_D m/s	ω_D ps	A_D ps ³	ρ	R_{Exp} ps ²	R_{Th} ps ²	Err %
MD-LJ	39.5	1320 [16]	540 [34]	2.1×10^{-2} [34]	0.120 [16]	1.14	610	6.99	8.8×10^{-3}	2.4	0.099	0.103	4
MD-SiO ₂	14.1	5940 [17]	3570 [17]	3.7×10^{-5} [17]	0.045 [17]	1.61	3950	63.7	1.2×10^{-5}	3.1	0.00055	0.00060	8
SiO ₂	15.1	5960 [9]	3750 [9]	5.3×10^{-5} [35]	0.042 [4]	1.58	4130	65.1	1.1×10^{-5}	4.9	0.00101	0.00053	64
GeO ₂	16.1	3650 [36]	2150 [36]	1.4×10^{-4} [37]	0.034 [37]	1.54	2380	36.8	6.0×10^{-5}	2.3	0.00237	0.00187	24
Glycerol	8.2	3600 [38]	1870 [38]	9.9×10^{-5} [39]	0.031 [40]	1.93	2090	40.4	4.5×10^{-5}	2.2	0.00177	0.00194	10
<i>o</i> TP	10.7	2940 [41]	1370 [41]	3.5×10^{-4} [41]	0.036 [40]	1.77	1540	27.3	1.5×10^{-4}	2.4	0.00445	0.00523	16
Selenium	30.0	1800 [42]	895 [42]	2.2×10^{-3} [43]	0.070 [44]	1.25	1000	12.6	1.5×10^{-3}	1.4	0.00935	0.02170	80

measured and predicted values. Overall, the agreement is very good, also in view of the approximation made in deriving Eq. (5) and, more important, of the experimental uncertainties associated with the determination of A and B .

In conclusion, we applied a recent theory of vibrational excitations in a disordered elastic medium [29] to the determination of the dynamic structure factor. This theory, that successfully explained the plateau in the T dependence of the thermal conductivity in glasses [29], is now found to explain one of the most intriguing features in the collective high-frequency vibrational dynamics of disordered systems: the k^2 dependence of the Brillouin linewidth, i.e., of the acoustic attenuation. The theory also predicts the existence of a relation between the excess DOS (the boson peak intensity) and the sound attenuation coefficient (the Brillouin peak width). A test of this prediction using existing experimental and simulation data gives an excellent outcome, thus giving further support to the validity of the theory itself. Finally, the theory also indicates that the transition between the k^4 law at very low frequency and the k^2 law for Γ_k [9] (open dots in the inset of 2) takes place at a frequency much lower than the boson peak frequency which, in turn, is much smaller than the Ioffe-Regel limit frequency.

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